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TABLE OF CONTENTS

ABSTRACT

The type of coupling between the pressure oscillation of an acoustic field and the flame zone of a solid propellant may be expected to depend on the pressure level and on the frequency of the acoustic field. Based on the granular diffusion flame model and on estimates of flame zone thickness derived from measured burning rates, a map can be drawn of three frequency regimes of acoustic interaction for a typical ammonium perchlorate propellant. It is postulated that, at the lowest frequencies (the so-call "zero-frequency" regime), a propellant burning under oscillating pressure should emit a gas stream of oscillating entropy, visible as a temperature wave carried with the stream. Waves that seemed to have some of the expected properties were observed photographically in 1960 by W. A. Wood of Rohm & Haas, Inc., using a plastisol-nitrocellulose propellant specimen.

In experiments at Princeton, a search was made for the predicted waves using a 2-inch diameter T-burner as the source of oscillating pressure, in order to confirm the observations of Wood, and then to measure the pertinent parameters, namely temperature, amplitude and phase. Unfortunately, no waves of the predicted type were found in a search that extended over the frequency range from 75 to 1000 cps and the pressure range from 250 to 1200 psi. The result was negative for nitrocellulose base propellants as well as for ammonium perchlorate composites. However, waves similar to those of Wood <u>were</u> observed reproducibly when burning was allowed on the sides of an uninhibited test sample, i.e., in a non-one-dimensional situation, but even so these waves were much weaker than the temperature variations that were expected. They have been explained as the result of fluctuating cooling effects in the annular cavity, having nothing to do with the flame zone of interest.

Various reasons for the non-occurrence of the expected temperature waves were considered, and some have been discussed in previous reports. The most likely explanation is that the model of the gas phase reaction zone originally assumed, namely, a thin one-stage reaction zone (50-100 μ thick) is partially in error. Instead, it appears that the gaseous zone may be composed of two portions - a thin primary zone near the surface needed in order to account for the observed burning rates, and a diffuse after - burning zone where the combustion reactions go to completion. Such a spread in the reaction zone would imply a longer total reaction time, thus introducing strong interaction phenomena that had been ruled out in the so-called "zero-frequency" description.

The conclusion to be drawn from this research is that the steady-state model used in the prediction of the temperature waves is partially in error. Further experiments at lower effective frequencies are being carried out to investigate the interaction with the flame zone and to determine the boundary of the "zero-frequency" regime.

INTRODUCTION

As detailed in earlier reports (1,2,3), the current study of solid propellant combustion instability has been focused on a determination of one part of the acoustic admittance of the burning surface, namely, the temperature-response function and its behavior as the mean pressure and the frequency of oscillation are varied. Most of the work has been done on ammonium perchlorate composite propellants, although nitrocellulose types have been tested for comparison.

Based on the granular diffusion model for the steady state burning zone, (4), three different pressure-frequency regimes of acoustic interaction have been laid out, as shown in Figure 1. The boundaries of these regimes are drawn using the criterion that, if the period of oscillation is long compared to the characteristic time of a component process, the process will be affected by the oscillation only in a quasi-steady manner. Specifically, a dimensionless time ratio can be defined by:

$$\tau = \frac{\text{process time}}{\text{period of oscillation}} \qquad (1)$$

For values of τ less than 1/10, the process can be said to behave in a quasi-steady manner, so this value may be used as an approximate criterion to separate the regimes of interaction of the pressure oscillation with the burning zone.

The two process times involved one in the gas, one in the solid, might be estimated as follows. Assuming a non-reactive composite solid with a one-stage gaseous reaction zone of granular diffusion type, the gas zone thickness turns out to be proportional to $p^{-1/3}$ at high pressure, and proportional to p^{-1} at low pressure. The gas velocity through the flame zone is proportional to $p^{-2/3}$ at high pressure and independent of p at low pressure. Dividing the gas zone thickness by the gas velocity, it is found that the characteristic time of the gaseous zone varies as $p^{1/3}$ at high pressure and p^{-1} at low pressure. A typical value at 250 psi, based on a gas efflux velocity of 250 cm/sec and a flame thickness of 50 μ, is 2×10^{-5} seconds. These considerations provide the basis for the top curve in Figure 1.

Consideration of the residence time of a propellant element in the solid phase of the flame zone provides the basis for the lower curve of Figure 1. The solid heat-up time varies as $p^{-2/3}$ at high pressure, and as p^{-2} at low pressure. At 250 psi, the time involved is 6×10^{-3} seconds, from a thickness of 30 μ and a burning rate of 0.5 cm/sec. (The effective depth of a thermal variation may be nearly an order of magnitude greater than this, and will increase with propellant diffusivity and decrease with increasing frequency).

In the area of high pressure and low frequency, the oscillation period is much longer than the overall combustion time of an element of propellant, taken from the moment it first senses the heat of the advancing flame in the solid state to the ultimate completion of reaction in the hot gas state. In this regime, the "zero-frequency" approximation holds, that is, the flame processes have sufficient time to adjust to any change in an absolute quantity or gradient, so that

steady-state conditions can be assumed to prevail at all times. The boundary of the zero-frequency regime is determined by the above-mentioned criterion that the period is ten times the over-all combustion time. For shorter oscillation periods (higher frequencies), it is useful to distinguish the two component parts of the combustion time, that is, the solid-phase heat-up time and the (very much) smaller gas-phase diffusion-reaction time. In the "low-frequency" regime, the gas phase can be treated in a quasi-steady fashion, but the solid phase must be treated dynamically. This case will be considered in some detail in a later section. In the "high-frequency" regime, the gas phase would have to be treated dynamically. The boundaries of these regimes are drawn in Figure 1 in accordance with the pressure and burning-rate effects on the characteristic times, as described above. The high-frequency regime could be further subdivided if we had an accurate knowledge of the times involved in the various component steps of the gas-phase combustion reactions. A fourth regime in Figure 1 can also be identified, when the pressure is sufficiently high and the flame layer sufficiently thin to make the simple one-dimensional granular diffusion flame model no longer admissible. There is as yet no theoretical description of this regime of combustion, although some research at Aerojet General Corp. has indicated that burning rates may be correlated with microscopic physical failures in the solid state. In any case, it is reasonable to set the high-pressure region apart as a fourth regime.

Cantrell, Hart, and McClure have recently drawn a similar map to consider the importance of the different flame processes. In their map, the coordinates were the burning rate and the frequency, but the same general arguments were used. The only differences were that the radiation feedback was considered important (although there is no strong evidence for this) and that a slightly different criterion was used to draw the boundary of the zone where a lag could exist in the solid heat-up zone.

A similar map for double-base propellants would probably be complex, because of the presence of more distinguishable flame steps, several being very strong functions of pressure and composition.

In the "zero-frequency" regime, two simple limiting cases can be considered for the entropy variation of the gas emerging from the flame zone of a solid propellant burning under varying pressure. These are: (1) the emergent gas is characterized by constant entropy during an oscillation, much like a sound wave reflection; (2) the flame temperature is constant, leading to entropy waves carried (i.e., convected) with burnt gases. Since the wavelength of these waves is determined by the flow velocity of the burnt gases (a few hundreds of centimeters per second), they will have a far shorter wavelength (millimeters - centimeters) than that of the sound-field, and these convection waves will be seen as a time and space-varying temperature field above the burning surface (Figure 2). The spatial distribution of temperature (or entropy) can be given approximately as

$$S_f(x,t) = \bar{S}_f + \epsilon \frac{\gamma-1}{\gamma} c_p \sin\left[\omega\left(t - \frac{x}{\bar{u}}\right) - \pi\right] \qquad (2a)$$

or

$$\frac{T_x}{\bar{T}_f} = 1 + \epsilon \frac{\gamma-1}{\gamma}\left\{2 \sin \frac{\omega x}{2\bar{u}} \cdot \cos\left(\omega t - \frac{\omega x}{2\bar{u}}\right)\right\} \tag{2b}$$

for a pressure fluctuation given by

$$P_f = \bar{P}_f (1 + \epsilon \sin \omega t) \tag{3}$$

By comparing (3) and either (2a) or (2b), it is seen that the temperature (or entropy) and pressure should be 180° out of phase, so that an observer stationed at the edge of the flame zone sees the entropy of the emerging hot gas oscillate sinusoidally with time, with high values of entropy emitted at low values of pressure.

Some encouragement for the correctness of this simple viewpoint was provided by Wood's report (5) of luminosity waves, seen by a streak camera, that were 180° out of phase with pressure for conditions of 800 psi mean pressure and 300 cps pressure oscillations. He also observed that going to higher frequency or lower pressure changed the phase of the luminosity wave, i.e., the luminosity led the pressure by less than 180°. The results were only photographic, so that the character of the waves could not be determined. However, the occurrence of such waves was taken as an indication that non-isentropic conditions could indeed exist at the edge of the flame zone. This burning zone-gas field interface is not strictly a fixed point in space relative to the solid surface, since it may be expected that the pressure fluctuation will change the flame stand-off distance. However, the measurements of temperature would be taken at distances x from the surface such that the mean-surface approximation would still be valid.

Further discussion of the points mentioned above can be found in the report by W. Waesche and M. Summerfield "Oscillatory Burning of Solid Rocket Propellants" (6). In addition, this report contains a discussion of the many processes involved in the problem of combustion instability, a review of the theoretical and experimental treatments of several of these processes, and an analysis of several candidate experiments for the investigation of one of these processes, namely, the coupling of a pressure oscillation with a surface flame zone. It was this analysis that led to the choice that a measurement of the temperature response function was the most promising diagnostic experiment that could be performed on the physics of this coupling.

EXPERIMENTAL EQUIPMENT

The experimental technique used for the observation of the predicted temperature waves has been described earlier (2). Briefly, it consists of a

T-burner (Figures 3,4), which functions as a source of oscillating pressure for an end-burning sample at one end of the T, and a radiometer consisting of a pair of collimating slits, an interference filter, and a phototube (Figure 5). By the addition of 0.2% NaCl, the propellant flame gases assume a blackbody character when viewed through this filter (owing to the high pressure and the substantial (2-inch) thickness of the flame), so that temperature variations can be measured simply by observing light intensity fluctuations with the phototube. The collimating slits insure that only the radiation from a thin plane, a known distance above the surface, is reaching the phototube. Since the sample surface recedes during a test, the radiometer record gives the temperature behavior of the gas field as a function of time and distance from the flame zone of the sample propellant.

In addition, photographic techniques were employed to observe and characterize the expected thermal waves. As seen in Figure 4, a Fastax camera was used for cinematography, while streak photography was also used to detect the waves. Full details are given in the previously-mentioned report (6), which also describes the operating characteristics and variables of the T-burner as a source of oscillating pressure.

EXPERIMENTAL RESULTS

Preliminary test firings, all at 430 cps, were made with a test propellant composed of ammonium perchlorate and an LP-3 binder. A sample containing 80% bimodal AP (70% 210μ - 30% 25μ) seemed to emit some bands at 300 psig with oscillations of 100 psig amplitude, but not at 600 psig with 170 psig amplitude. In addition, the same basic composition, but with a 45μ - 5μ oxidizer distribution, did not emit any detectable thermal waves at either 350 psig or 650 psig. In an attempt to make the waves more detectable, 0.1% NaCl was added to the LP-3 propellants, but no evidence of waves was obtained.

The first propellant system to be studied extensively under oscillating pressure conditions was the polyurethane system. Observations were made at frequencies from 77 cycles per second to 950 cycles per second.

The photographic (color) results for end-burning samples containing 85% trimodal ammonium perchlorate are shown in Table I below.

TABLE I

Observation of Luminosity Waves from Polyurethane Propellant Containing 85% Ammonium Perchlorate.

Frequency (cps)	Pressure (psig)	Amplitude (psig)	Remarks
950	600	15	No waves
430	430	150	Narrow faint band emitted from surface about 90° after a pressure minimum.
265	330	85	
190	270	80	Wide faint band emitted from
150	310	50	surface near pressure minimum.
105	290	90	
77	455	80	

In no cases were these waves as distinct as had been expected, and determination of phase relations was quite difficult.

The effect of oxidizer particle size was studied by using a finer perchlorate blend, (70% 45 μ - 30% 5 μ); which would lead to a thinner combustion zone. Because of the excessively high viscosity of uncured propellant resulting from the finer particles, the oxidizer content was lowered to 80%. The photographic (color) results for the 80% - 20% propellant are given in Table II below.

TABLE II

Observation of Luminosity Waves from Polyurethane Propellant Containing 80% Ammonium Perchlorate

Frequency (cps)	Pressure (psig)	Amplitude (psig)	Remarks
950	600	15	
430	720	120	
	400	180	No Waves
	310	150	
265	1150	90	
	850	95	
265	330	150	
190	450	55	Wide faint bands, becoming more
150	340	75	diffuse as frequency dropped.

No particular difference in behavior was observed for the different oxidizer sizes, although the bimodal grind samples did seem to emit slightly clearer waves. As a result, propellant based on the bimodal perchlorate was chosen as the base propellant for addition of 0.1% salt, both to intensify the waves for photographic observation and to enable the use of the luminosity equipment. Further tests were then made for the conditions discussed in Table III.

TABLE III

Conditions for Observation of Luminosity Waves from Polyurethane Propellant with 0.1% Salt

Frequency (cps)	Pressure (psig)	Amplitude (psig)
950	600	10
430	510	55
	430	150
	340	140
265	840	75
	570	65
	330	105
190	1060	125
	250	85

Neither the phototube radiometer nor the film record (black-and-white through a Na filter) showed any entropy variation. The fluctuation of luminosity recorded by the phototube was exactly in phase with the pressure, and the film record showed that the entire visible field had a uniform emissive power, although varying sinusoidally with time (Figure 6).

Various explanations for the unexpected lack of distinct thermal waves were advanced. The first concerned the possibility that one driver grain was too close to the sample surface, and its combustion gas was interfering with the gas flow from the sample surface, through back flow and turbulent mixing, thus "poisoning" the entropy waves and resulting in the cancellation of wave effects. To reduce this, a 4 in. spacer assembly was added, giving a distance of 6 in. between the inhibited end of the driver grain and the test sample surface. A series of runs was made using polyurethane samples, with the results shown in Table IV.

TABLE IV

Conditions for Testing Standoff Effect with Polyurethane Samples

Frequency (cps)	Pressure (psig)	Amplitude (psig)	% AP	Remarks
400	350	95	85	Narrow faint band emitted from surface about 90° after pressure minimum.
255	700	75	80	Diffuse bands about 45° after minimum.
	560	80		
	480	70		
115	620	70	80	Diffuse bands, too weak to obtain phase.
	560	70	85	

Comparison of the results in Table IV with those of Tables I and II indicates that driver proximity had no effect on the lack of emission of thermal waves. In fact, no difference in the appearance of the gas zone in the sample chamber could be detected, indicating that the amount of backflow and turbulence from the driver grain was minimal. The flow from the surface appeared absolutely straight and almost non-turbulent.

Another possibility was based on the thought that the sodium added to the propellant to bring out the thermal waves might, in fact, be smearing them out by some unidentified means. The possibility of a high concentration effect causing an obscuration of all but the near boundary layer was also considered. To test these possibilities, the salt concentration was varied from 0.02% to 0.5% NaCl and photographs and radiometric traces were taken for a wide range of pressures and frequencies. Color movies were taken for the lower concentrations, and black-and-white movies were taken through a sodium filter for the higher concentrations. In no case were thermal waves observed.

The method of adding the sodium to the test sample was also considered. To make sure that bunching of NaCl particles was not occurring, the salt was

ground in a grinder at 12000 RPM, screened, and kept dry in an oven until placed in the mixer hopper. Microscopic observation of the propellant surface did not show any salt particles larger than 10 - 20 μ. One difficulty was never overcome, however. The sodium was always added as a salt which was insoluble in the polyurethane binder. Attempts to use sodium compounds which would dissolve in the polyurethane were unsuccessful, since all compounds tried affected the polymerization of the fuel adversely.

All this concern over the sodium seemed beside the point, however, considering the previously mentioned fact that Wood (5) had photographed clear luminous waves emitted from the surface of a plastisol-nitrocellulose type propellant containing ammonium perchlorate. In his tests, Wood did not have to add sodium at all, to make the waves visible.

As a result, next consideration was given to possible differences in combustion characteristics between the plastisol system used by Wood and the pure composite systems used in the original (although exhaustive) tests performed here. At about the same time, facilities for making plastisol-type propellants had been set up in the Solid Propellant Processing Laboratory of the Guggenheim Labs through the kind assistance of experts of the Rohm & Haas Company's Redstone Arsenal Research Division, who had developed the processing techniques and made the propellants used by Wood.

A series of tests followed with plastisol propellants. Full compositional details are given in Appendix A, so only distinguishing characteristics will be given. Test conditions were as given in Table V.

TABLE V

Test Conditions for Plastisol Propellant Samples

Frequency (cps)	Pressure (psig)	Amplitude (psig)	Propellant
255	650	70	P-4
	620	90	P-6
	700	60	
	600	55	P-7
	700	50	
	680	40	P-8
	550	50	
	570	60	P-9
180	850	45	9a-67
	900	50	
	700	40	
	910	45	P-7
	700	40	
175	700	50	9a-67

It should be noted that the runs were generally at higher pressures than for the polyurethane. Spacers were used in every case, both to insure no backflow and to

give amplitudes comparable with those used by Wood, which were of the order of 75 psi for the 800 psi firings. The distinguishing characteristics of the plastisols made here, all designated with a P-No., are as follows. All contained 35% AP, which was 70% 45μ - 30% 5μ, except for P-6, which contained all 5μ perchlorate. Batch P-6 also contained 0.2% NaCl, while P-8 was identical to P-4 plus 0.2% NaCl. Batches P-4 and P-7 were identical, as were P-8 and P-9. The 9a-67 propellant was furnished by Rohm & Haas, and was the same type as that used by Wood in some tests.

The results of the firings were quite bewildering, since no waves of the type reported by Wood were observed, either photographically from movies or streak photographs or radiometrically. In every case, more than one run was made for a set of test conditions. The same statement can be made for the different tests reported with the polyurethanes, where duplicate runs were made, first with black-and-white film, to establish proper exposure, then with color film.

A search for minor compositional differences between propellants used at Princeton and those used at Rohm & Haas was carried out. Although exact formulations were classified, it was found that a slightly different ball powder (containing no carbon black) was used, no stabilizing agent was added, and that the anti-caking agent used on the ammonium perchlorate was magnesium oxide, rather than the tri-calcium phosphate present on the oxidizer as received at Princeton. Successive batches made here eliminated the stabilizer (P-10), changed the ball powder (P-15), and changed the oxidizer particle size (P-16). Firings were made over a range of pressures at oscillation frequencies of 180 and 255 cycles per second, but no waves were photographed at all.

The next avenue investigated was the configuration of the test sample. Information received from Dr. Wood (7) revealed that the disk thickness was 0.25 inches, while the sample disk diameter was 1.50 inches in a cylindrical chamber having a 1.85 inch diameter. It was further revealed that waves had been observed only in tests where the sample had been left uninhibited to insure that the windows were not dirtied, and that a few inhibited firings, late in his test program, had shown no waves. The lack of observable waves in those tests was ascribed by Wood to experimental difficulties.

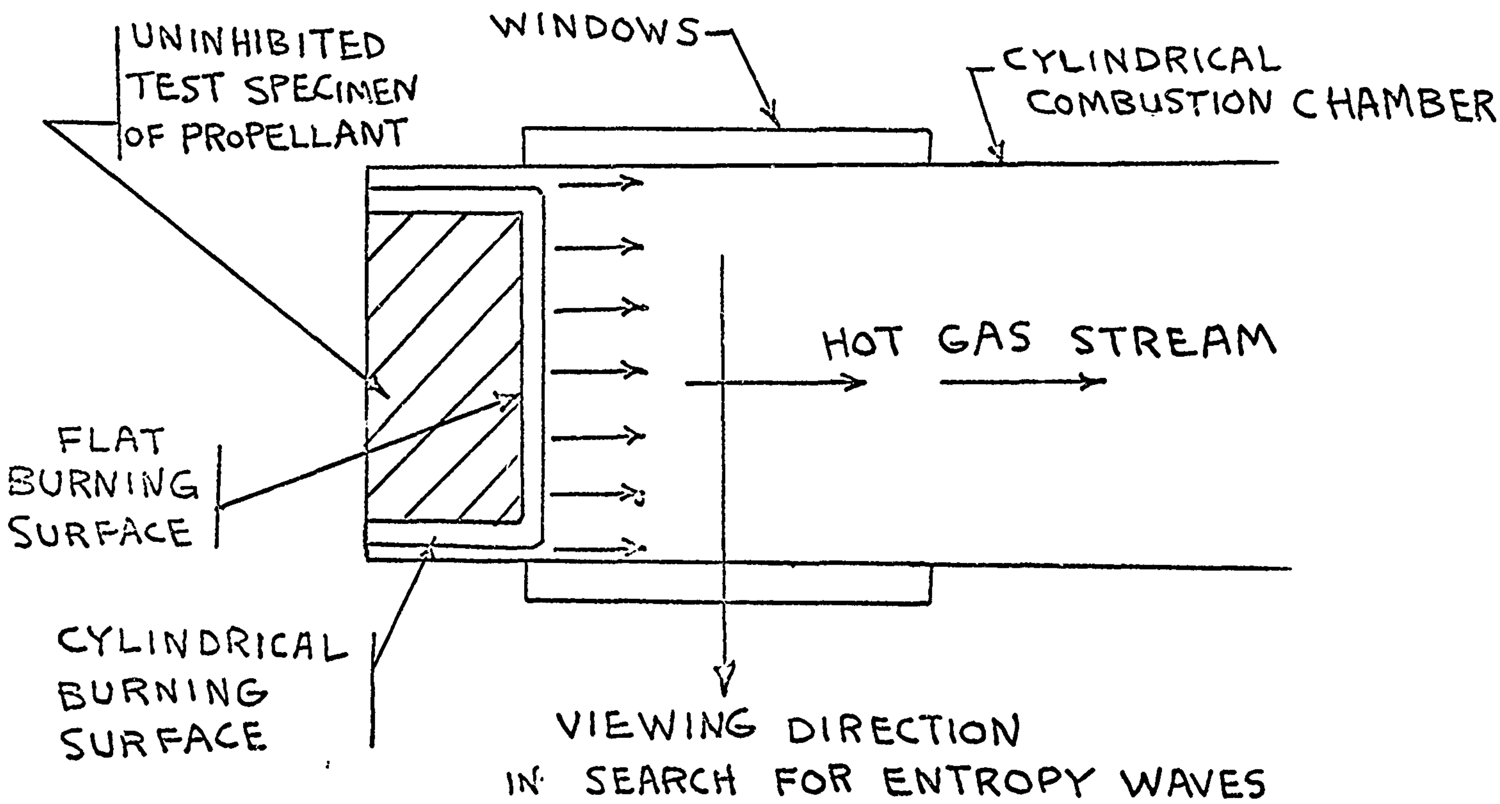
As a result of this communication, a series of firings was conducted. Duplicate conditions were used, except that the sample was uninhibited in one firing and inhibited in the other. To insure window clarity and even burning, the inhibited samples were cast into steel cups and cured. Although the surface was blocked from view, the observations of interest were away from the surface, so the cup technique was felt to be a good one for this series. The sets of conditions given in Table VI were used.

TABLE VI

Test Conditions for Comparing Inhibited and Uninhibited Test Samples

Frequency (cps)	Pressure (psig)	Amplitude (psig)	Propellant
255	760	45	P-11
	850	30	
	1040	40	
175	880	55	P-11
	700	40	9a-67
	780	40	P-15
	640	35	
	770	40	P-16
	650	35	
	770	40	
			85% trimodal AP, 15% polyurethane
	650	35	
	790	35	80% bimodal, AP, 20% polyurethane

In every one of these twelve sets of firing conditions, luminous waves were emitted from the propellant when the sample was uninhibited, while the inhibited samples gave no waves, indicating that the luminous waves previously observed by Wood (5) did not arise from a one-dimensional situation. (When the inhibitor is omitted from the cylindrical side surface of the sample propellant disk, burning can take place in the annular chamber formed between the cylindrical propellant surface and the inner surface of the firing chamber. The combustion gas emerging from the zone can be seen to form a cylindrical sheath around the main stream of gas emerging from the flat face. See sketch.)



The waves were observed both by colored movies and by black-and-white streak photographs (Figures 6,7,8). Sample diameters of 1.50 in. and 1.75 in. were used in the Princeton test chamber, which has a diameter of 1.938 in., and 0.25 in. thick samples were used. No definite statement can be made as to the comparative strength of the waves for the different sample diameters used here.

Possible explanations for the nonoccurrence of entropy waves in a one-dimensional situation will be considered in the following sections. Some preliminary tests were performed to characterize the waves found at Princeton for the uninhibited (non one-dimensional) case. These tests involved the addition of salt to the test sample to see if the waves were strictly temperature waves. However, streak pictures taken through a sodium D-line filter indicated very weak wave behavior, especially after equilibrium pressure (600 psig) had been reached. In addition, simultaneous radiometric traces, also through a sodium D-line filter, revealed that the luminosity fluctuations were mainly in phase with the pressure. Both of these results indicated that one-dimensional entropy waves still did not exist in the main stream.

The most logical explanation for the generation of waves from the uninhibited, side-burning test specimen is involved with wall effects, owing to the large heat sink represented by the cooler walls. The pressure fluctuations lead to periodic cooling of the gases, resulting in the liberation of temperature waves in the thin annular stream. Therefore, these waves are a type of entropy wave, but they result from an oscillatory cooling effect.

This mechanism of wave generation also explains the failure to observe the waves with salted propellant. The dominant radiation from the salted test speciment comes from the center of the specimen, and would overwhelm any variations in luminosity from the edge. In the case of unsalted propellants, however, the gases emitted from the edges would be more visible. Systematic tests with varying sample diameter, and with salt only in the outer portion of the test specimen, should help to reveal the magnitude of any such periodic cooling effects.

POSSIBLE SOURCES OF DISCREPANCIES

The failure to observe the predicted entropy waves in a one-dimensional situation leads to a study of possible mechanism through which the waves can be damped out or reduced in magnitude. First of all, possible inconsistencies in the experiment must be considered.

Calculations have been reported earlier for the damping of thermal waves by streamwise heat conduction in the gas phase (1). In these calculations, it was shown that waves should not be seriously damped by thermal conduction for low frequencies and high pressures (i.e. long wavelength and low thermal diffusivity in the gas), precisely the conditions under which most of these experiments were conducted. In addition, the weak luminous waves observed in the tests in which annular bruning was allowed did not show signs of such decay.

The appearance of the waves further negates two other possible mechanisms by which the predicted waves could be damped, i.e., acoustic streaming and mixing

phenomena. The planar nature of the observed waves indicates that they are not being affected by any motion in the gas field other than the purely one-dimensional type associated with the acoustic field. In addition, neither movies nor streak camera records showed any indication of swirling or turbulent gas motion which could have disrupted waves of the expected type.

Since there appear to be no obvious experimental reasons for the failure to observe the strong thermal waves predicted, the physical model of the flame on which their appearance was based must be examined more closely. In an earlier report (3), calculations were made which showed that the boundary on the basis of physical intuition between the "zero-frequency" and "low-frequency" regimes was consistent with the calculated on the basis that the longest time lag in the combustion process occurred in the solid phase through heat conduction. These calculations showed further that such a non-steady thermal lag could cause some flame temperature fluctuation at relatively low frequencies, and these fluctuations would act to weaken the expected entropy waves in the "low-frequency" regime of acoustic interaction.

Because of differences in the results of calculations made here and those reported by Hart and Cantrell (8), possible differences in the models used were considered. The major difference lay in the fact that the sensitivity of burning rate to the thermal gradient at the solid surface was taken into account by Hart and Cantrell. Accordingly, the model used here was extended to include this sensitivity, but it was found that the effect was slight for values of $\omega\tau_s < 10$, which is the region of most interest. (A fuller account is available in (6)).

Another factor that could affect the interaction frequency regime would be a thickening of the combustion zone and a consequent increase in the transit time of a particle through the combustion zone. The most likely cause for a possible error in estimating transit times through the reaction zone is the structure of the gaseous phase of the flame zone. For a heat-and mass-transfer balance to exist in steady-state combustion, there must be a steep temperature gradient immediately adjacent to the surface. This gradient could be caused by the occurrence of the bulk of combustion in a thin zone (of the order of 100 microns) near the surface. However, it is quite possible that this concentrated combustion zone is followed by a rather diffuse after-burning zone where the combustion reactions go to completion. Such an after-burning zone would modify the pressure-flame interaction in two ways. First, the transit time could be affected, and second, a corollary, the thermochemistry could be affected. If, for instance, pressure-sensitive reactions were involved in the completion of combustion, the energy release could fluctuate as a function of time at the edge of the flame zone.

The determination of the thickness of the flame zone is not a simple matter. Ordinary photographs do not suffice. Some qualitative experiments conducted here indicate, however, that the total flame zone thickness may be of the order of 1 - 2 millimeters, rather than 100 microns, as reported by Summerfield (4). Sabadell (13) observed that thermocouple traces leveled off in the gas phase of the flame for 2 millimeters or more at pressures as high as 1000 psig before rising a second time, which shows that, for some propellants, the gaseous reactions may occur in two stages.

Another set of experiments concerned the spectral distribution of the flame zone. Full details of emitters, experimental layout, etc., are given in (6). The significant feature of the experiments was that CN radiation (the 3883 Angstrom band) was not confined to the surface region but extended, in some cases, over several millimeters from the surface. These were based on exposures of 1/10 second and viewing the relative strength of spectral lines at different heights above the surface, since the burning surface was perpendicular to the slit in these tests. During this time, the burning surface would move about 1 millimeter, depending on the burning rate. Another source of error was the finite thickness of the burning sample, since the cone subtended by the quartz lens used to focus the flame on the spectrograph slit had a height of nearly 1 millimeter at the edge of the sample. Adding these to the aberrations of the simple lens system and the viewing windows, it is difficult to say how exact quantitative measurements can be made with this particular experimental arrangement. It was observed, however, that a mercury arc used for backlighting showed a fairly sharp cutoff at the surface. The observation that the CN bands were not confined to the surface region, but extended, in some cases, over more than 3 millimeters from the surface, indicates the possibility of a reaction zone with a thickness of 1 - 2 millimeters at the pressures used (up to 30 atmospheres).

More exact tests are being carried out by Povinelli (14) and Selzer(15), who are determining quantitative distribution of emitters through the reaction zone. Their results indicate that appreciable emission takes place up to at least 2 millimeters from the surface, although both are working at atmospheric pressure. The results of Watermeier (16) indicate a reaction zone thickness of .8 - 3 millimeters at pressures from 250 to 750 psi, and Penzias(17) found, using an infrared technique, that the flame temperature was not attained until at least 1 millimeter from the surface at a pressure of 800 psi. In no case was a narrow cut of oxidizer used, so that some of the uncertainty could have resulted from different crystal sizes. In addition, the effect of pressure on apparent reaction zone thickness has not, as yet, been carefully studied.

The effect of these experiments is, however, to cast substantial doubt on the thickness originally assumed (50μ) for the gas phase reaction zone, and to give strong indications that a more diffuse region may exist which could be affected in a number of different ways by fluctuating pressure.

CONCLUSIONS AND FUTURE PLANS

The conclusion to be drawn from the experiments reported here is that the commonly accepted model of the solid-propellant flame zone needs modification. The most likely portion needing revision is the structure of the gaseous portion of the reaction. In addition, it has been shown that the luminous waves reported by Wood and thought to provide evidence for the existence of a thin flame zone were probably caused by the particular configuration of his burner, and are not the result of a truly one-dimensional pressure-flame interaction.

The search for the boundary of the "zero-frequency" regime is continuing since this regime must exist in the steady-state limit. Two approaches are being used; they are as follows. First the sensitivity of the experiment is being refined. More quantitative photographic techniques will be employed to record any possible faint waves that might be emitted by samples burning under oscillating pressure, using the same T-burner apparatus as used up to this point. Second,

the effective frequency range is being lowered, i.e., a given fractional change in pressure is being extended over a longer time. The lower value of dp/dt used thus far has been about 20,000 psi/sec, corresponding to an 80 psi amplitude oscillation with a frequency of 77 cps. Lowering this value, while keeping the relative pressure variation constant, cannot be accomplished with the present apparatus, since the frequency range is limited by the size of the test cell and practical considerations such as heat loss. As a result, the experiment shown in Figure 9 has been set up. In this experiment, the pressure in the chamber is varied by moving the plunger in the nozzle. At the same time, the temperature of the flame gases is measured by the radiometer seen in Figure 9. For some value of dp/dt, the flame temperature should remain nearly constant, as expected in the steady-state limit. This value of dp/dt will define the boundary of the "zero-frequency" regime for the particular propellant under test. Above this value of dp/dt we might expect to observe a positive value of $dT_{flame}/dp_{combustion}$, to correspond with the constancy of entropy with time, as seen in the oscillator experiments. The variation of this limiting value of dT_f/dp_c for different propellant and test variables should give valuable information for the treatment of steady-state and unsteady-state burning.

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LIST OF SYMBOLS

c_p Specific heat of burned gas

p Pressure of burned gas

s Entropy/unit mass of burned gas

t Time

T Temperature of burned gas

u Velocity of burned gas leaving flame

x Distance from flame

γ Ratio of specific heats

ϵ Fractional pressure variation

τ Characteristic time of combustion

ω Frequency of oscillation, rad/sec

Superscript

$\bar{\ }$ Time-average value

Subscripts

c Combustion

f Evaluated at outer edge of flame

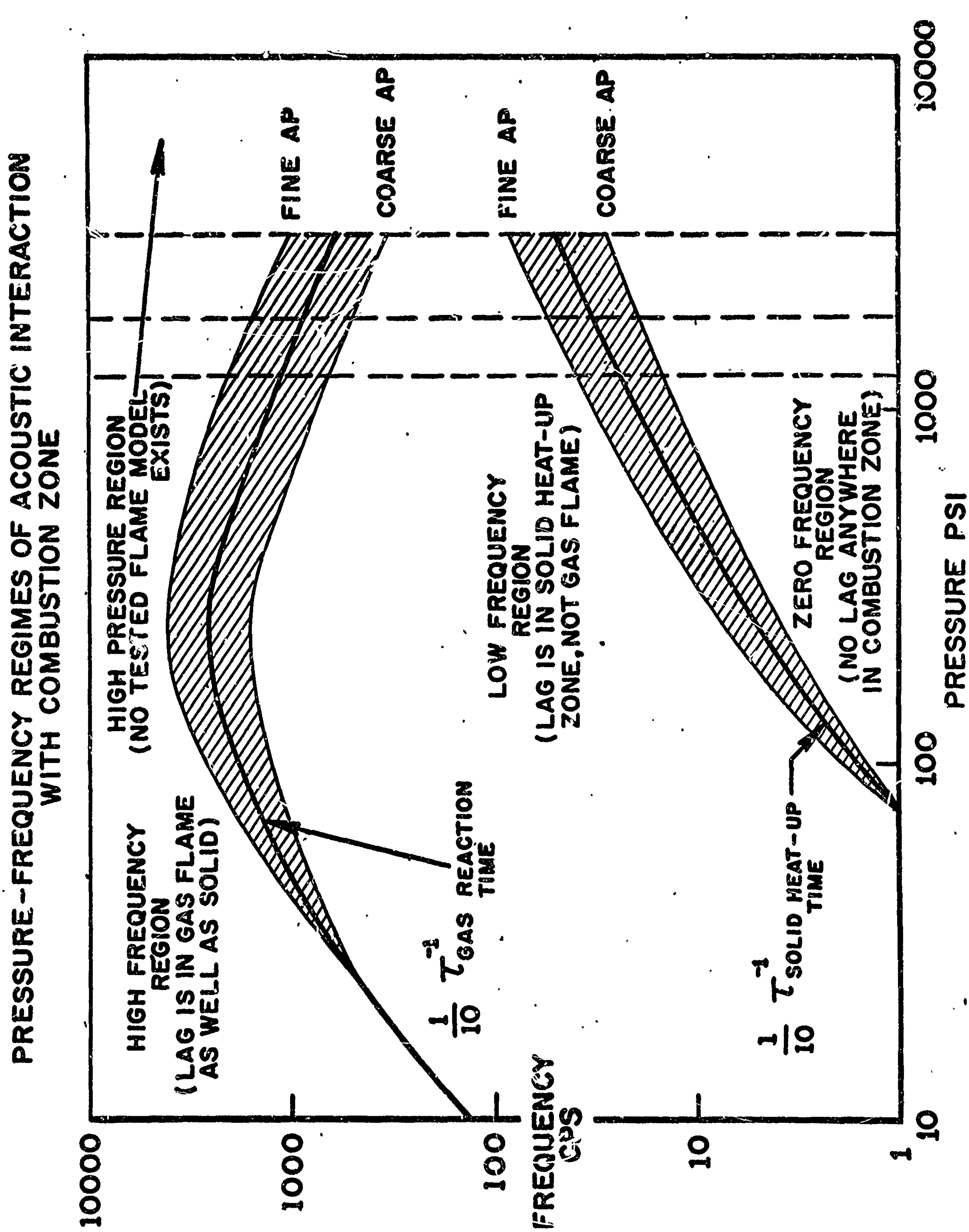
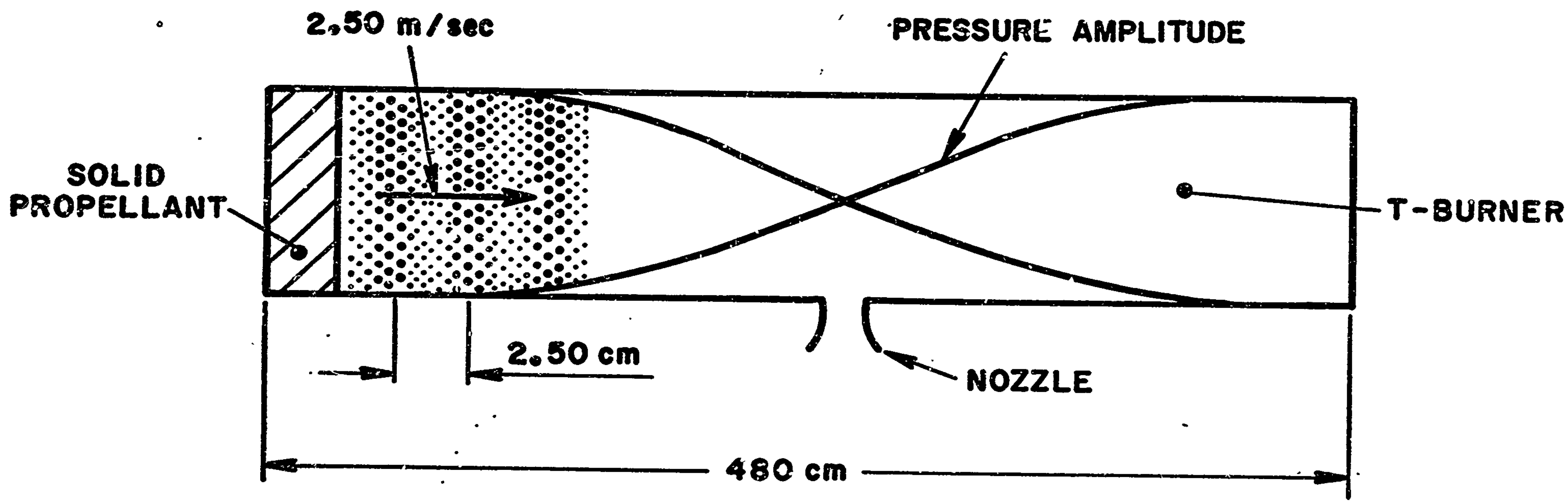

PRESSURE-FREQUENCY REGIMES OF ACOUSTIC INTERACTION WITH COMBUSTION ZONE
HIGH PRESSURE REGION (NO TESTED FLAME MODEL EXISTS)
HIGH FREQUENCY REGION (LAG IS IN GAS FLAME AS WELL AS SOLID)
LOW FREQUENCY REGION (LAG IS IN SOLID HEAT-UP ZONE, NOT GAS FLAME)
ZERO FREQUENCY REGION (NO LAG ANYWHERE IN COMBUSTION ZONE)
FINE AP
COARSE AP
FINE AP
COARSE AP
$\frac{1}{10}\ \tau_{GAS}^{-1}$
REACTION TIME
$\frac{1}{10}\ \tau_{SOLID\ HEAT-UP}^{-1}$
TIME
FREQUENCY CPS
10000
1000
100
10
1
PRESSURE PSI
10
100
1000
10000

FIGURE 1

ENTROPY WAVE PROPERTIES IN AN ACOUSTIC FIELD



	ENTROPY WAVES	ACOUSTIC WAVES
FREQUENCY	100 cps	100 cps
VELOCITY	2.50 m/sec	960 m/sec
WAVE LENGTH	2.50 cm	960 cm

FIGURE 2

JPR 2200

CROSS - SECTION OF T - BURNER

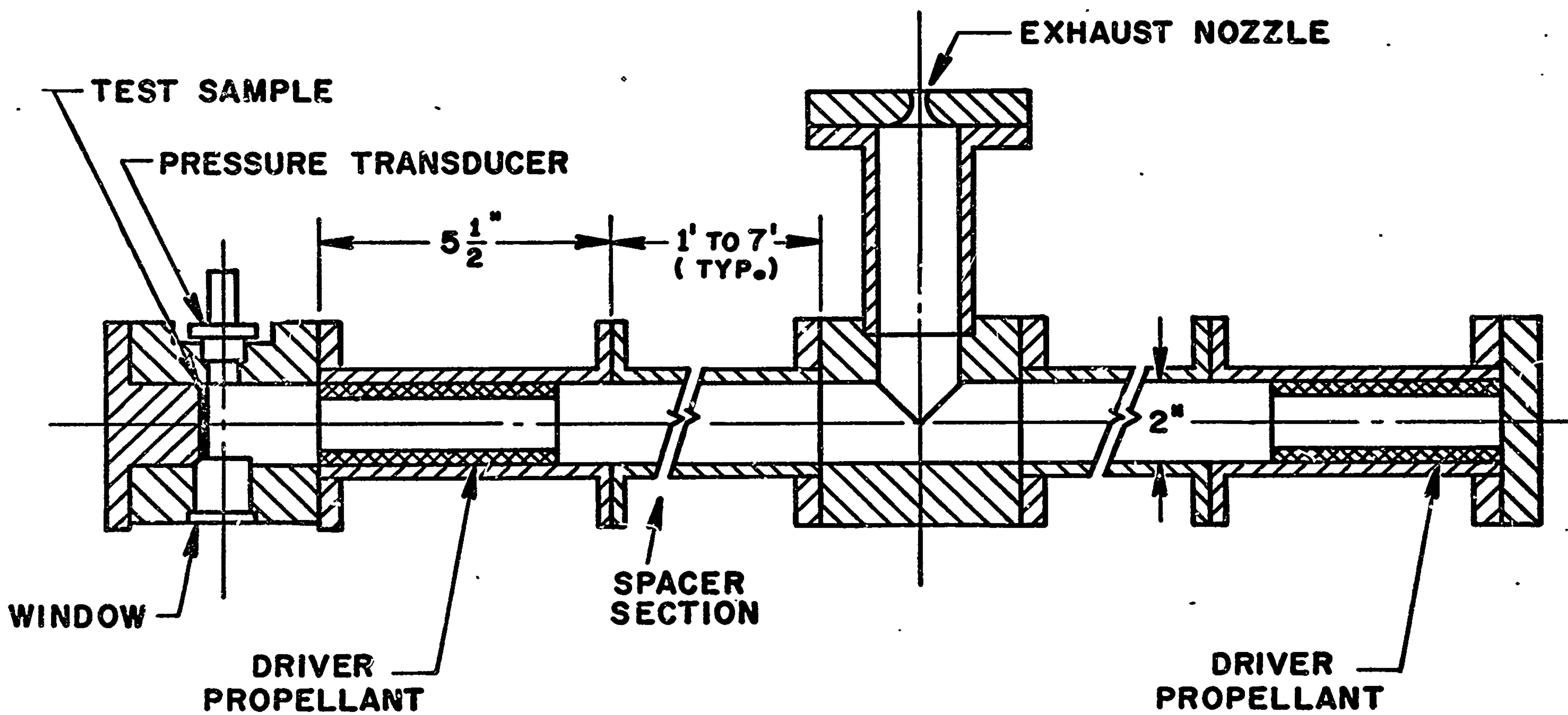


FIGURE 3

EXPERIMENTAL ARRANGEMENT FOR OBSERVATION OF TEMPERATURE WAVES
UNDER OSCILLATING PRESSURE IN T-BURNER

Figure 4

PHOTOMETRIC TEMPERATURE MEASURING APPARATUS

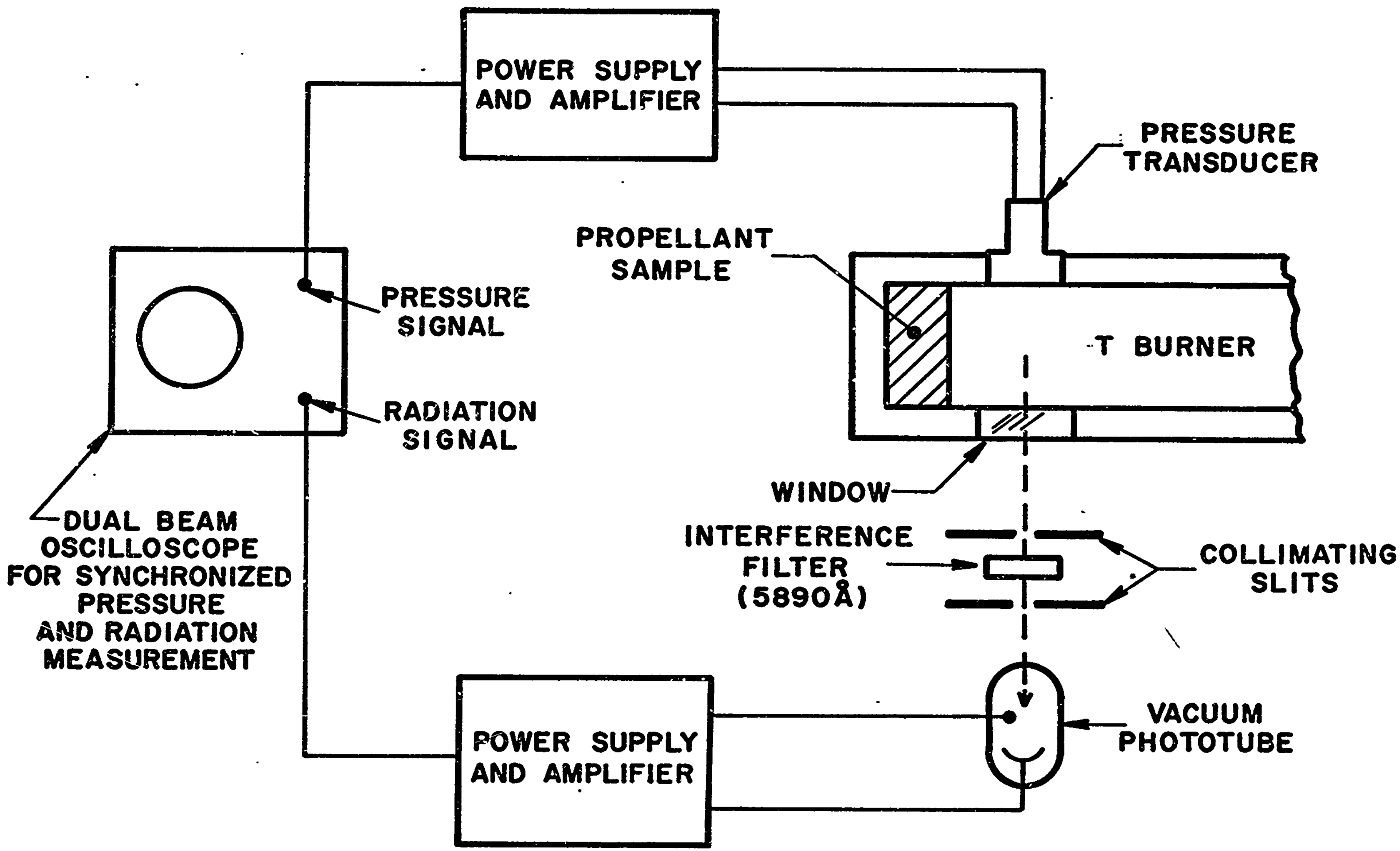
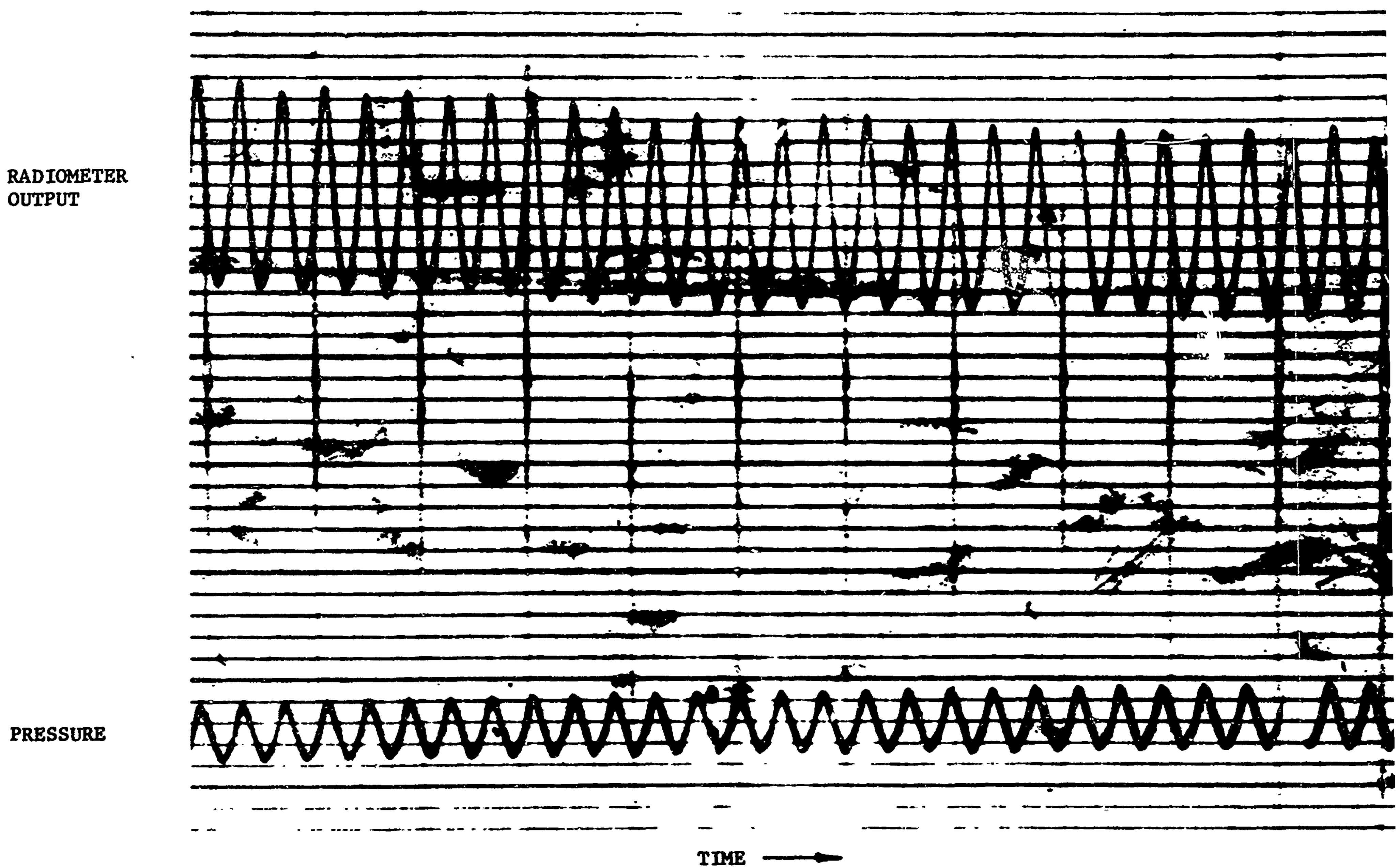
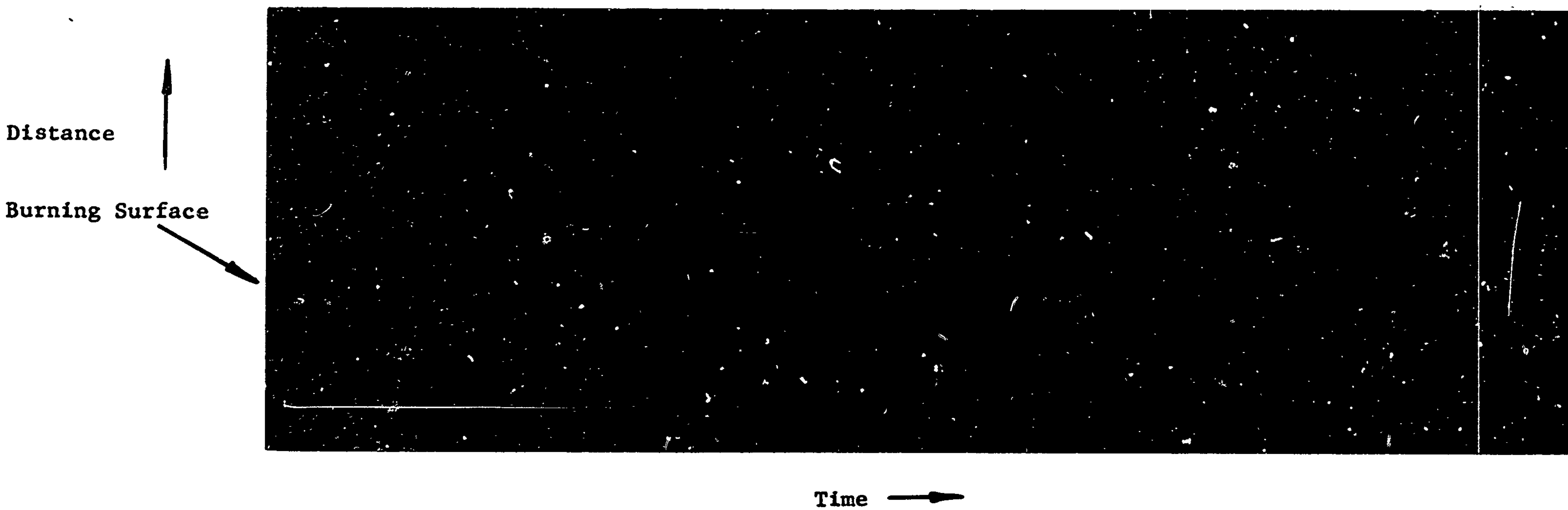


FIGURE 5



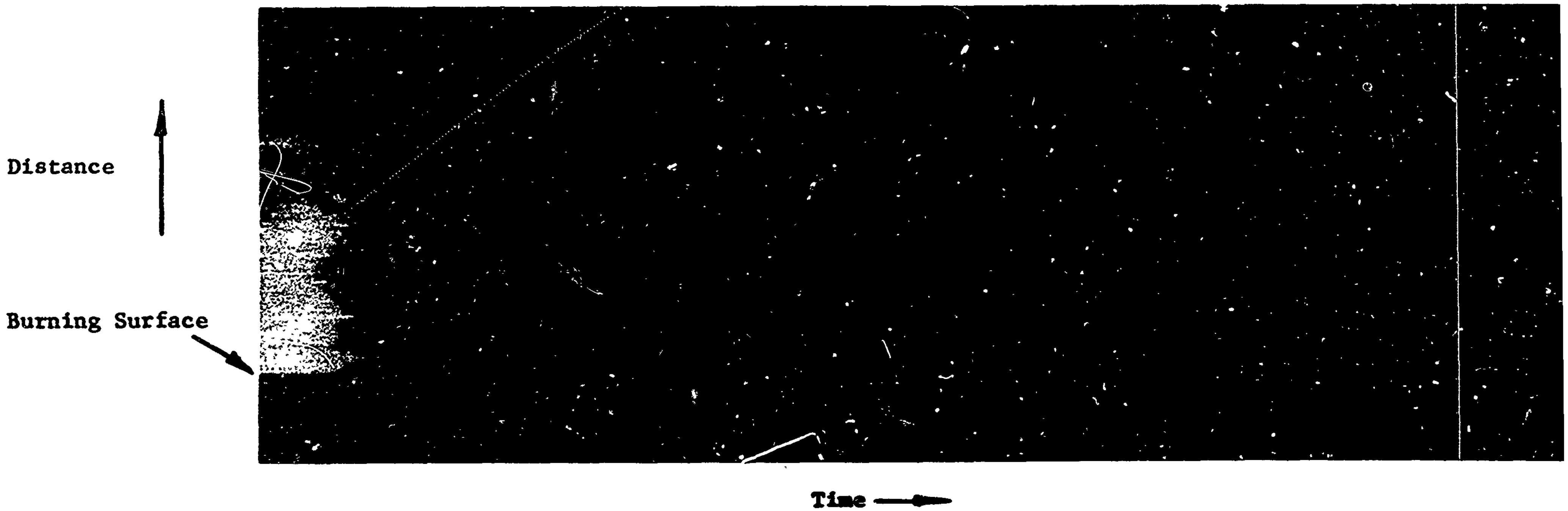
PORTION OF THE OSCILLOGRAPH RECORD OF RADIOMETER AND PRESSURE OUTPUT

FIGURE 6



STREAK CAMERA RECORD FOR AN INHIBITED SAMPLE
(Princeton)

FIGURE 7



STREAK CAMERA RECORD FOR UNINHIBITED SAMPLE
(Princeton)

FIGURE 8

CONFIGURATION OF DYNAMIC COMBUSTION EXPERIMENT

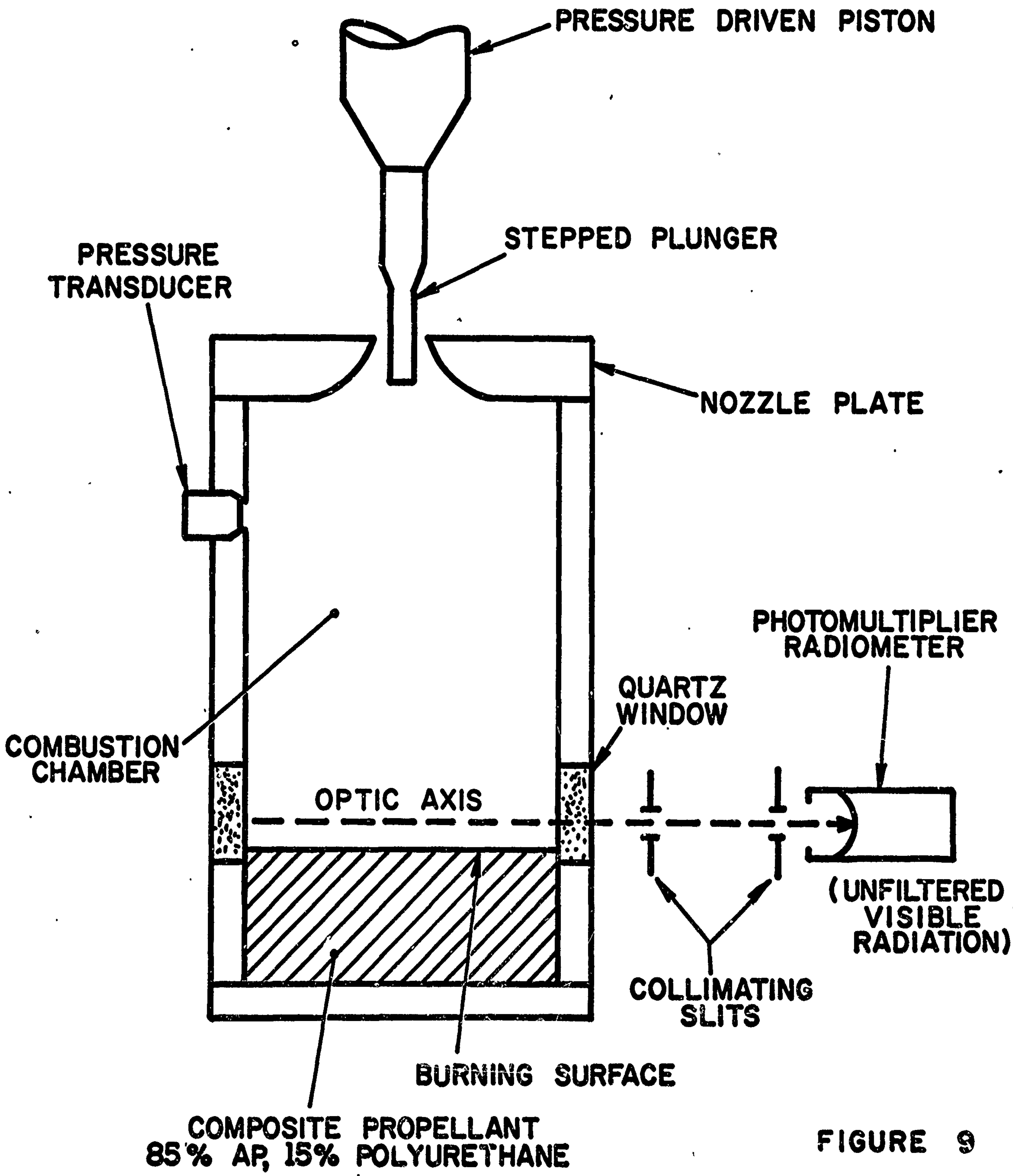


FIGURE 9

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13. ABSTRACT The type of coupling between the pressure oscillation of an acoustic field and the flame zone of a solid propellant may be expected to depend on the pressure level and on the frequency of the acoustic field. Based on the granular diffusion flame model and on estimates of flame zone thickness derived from measured burning rates, a map can be drawn of three frequency regimes of acoustic interaction for a typical ammonium perchlorate propellant. It is postulated that, at the lowest frequencies (the so-call "zero-frequency" regime), a propellant burning under oscillating pressure should emit a gas stream of oscillating entropy, visible as a temperature wave carried with the stream.

In experiments at Princeton, a search was made for the predicted waves using a 2-inch diameter T-burner as the source of oscillating pressure, in order to confirm the observations of Wood, and then to measure the pertinent parameters, namely temperature, amplitude and phase. Unfortunately, no waves of the predicted type were found in a search that extended over the frequency range from 75 to 1000 cps and the pressure range from 250 to 1200 psi. The result was negative for nitrocellulose base propellants as well as for ammonium perchlorate composites. However, waves similar to those of Wood were observed reproducibly when burning was allowed on the sides of an uninhibited test sample, i.e., in a non-one-dimensional situation but even so these waves were much weaker than the temperature variations that were expected. Various reasons for the non-occurrence of the expected temperature waves were considered, and some have been discussed in previous reports. The conclusion to be drawn from this research is that the steady-state model used in the prediction of the temperature waves is partially in error.

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